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PINE-OIL AND PINE-DISTILLATE PRODUCT EMULSIONS: METHOD OF PRODUCTION, CHEMICAL PROPERTIES, AND DISINFECTANT ACTION.

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PURPOSE OF INVESTIGATION.

The use of a pine-oil emulsion made from "steam" or "steam-and-solvent" process pine oil was advocated by Stevenson (18),[1] of the Hygienic Laboratory of the United States Public Health Service, in 1915, for general disinfecting purposes. He stated that such a product has a Hygienic Laboratory phenol coefficient of 4 to 6, which "remains practically constant for about two months. After that a noticeable deterioration occurs. Samples four months old show a phenol coefficient of about 3.5." According to McCoy, Stimson, and Hasseltine (13), "Hygienic Laboratory pine-oil disinfectant * * * is well adapted to the disinfection of intestinal discharges, but appears to be deficient against pyogenic organisms," and "in the present state of our knowledge the use of this preparation should be confined to bacillary infections such as typhoid fever, cholera, and bacillary dysentery." In October, 1917, Walters (31) pointed out the fact that pine oil is very much less active than phenol against the *Staphylococcus aureus*.

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[1] The figures in parentheses refer to the bibliography at the end of this bulletin.

Pine-oil products of the composition recommended by the Hygienic Laboratory and of similar composition have been put on the market by a number of manufacturers. Many of the commercial so-called pine-oil disinfectants are adulterated with kerosene and other mineral oils, and their phenol coefficients fall far below that found by Stevenson (18) for the Hygienic Laboratory pine-oil disinfectant.

The work reported in this bulletin was undertaken for the purpose of determining the physical, chemical, and disinfectant properties of pine-oil and other pine-distillation products in order to secure data to assist in the detection of the adulteration of commercial products, as well as to check up the statements concerning the deterioration of pine-oil disinfectant and its peculiar behavior against certain pathogenic organisms.

PRODUCTION OF PINE OIL.

Pine oil is obtained by the distillation of pine wood in closed retorts. It is an essential oil the odor of which varies from a pleasant pine to a disagreeable empyreumatic, depending on the method of its manufacture and the refining processes used. There are two general processes for making it—destructive distillation and the so-called steam or steam-and-solvent method. The wood used is the same in either case.

In the early days what was considered to be a vast, inexhaustible pine forest, beginning in North Carolina, extended down the coast through Florida and the Gulf States to eastern Texas. Soon the turpentine and lumber industries took most of the standing timber, and they have moved steadily south and westward from North Carolina, where they started, until they have almost reached the end of the forest in Louisiana and Texas. These industries left in their wake a large amount of waste in the form of stumps and dead and down timber. In the course of time, the outer layer rots away from this timber, and the resinous heart material remains, forming what is termed throughout the South "lightwood," so called because it is used to make fires and for lights, not on account of its density. As a matter of fact, it is heavy, some samples having a density as high as 1.075 (30). This is the wood which is used for distillation. Most of it comes from the longleaf southern pine (*Pinus palustris*), although commercially any wood that is "fat" enough, or contains enough resinous material, is used. This includes small amounts of slash pine (*Pinus heterophylla*) and shortleaf pine (*Pinus echinata*) (8) (19). Much of the wood is obtained in the course of clearing land for agricultural purposes.

DESTRUCTIVE DISTILLATION.

The crude beginnings of destructive distillation are to be found in charcoal burning (27), which was carried out in open trenches, the combustion being controlled by partially covering the wood with earth. Tar and charcoal were the only products recovered. Then came the beehive oven, operated in much the same way, which recovered the more volatile parts of the distillate.

The first retort plant, started about 1872 (14) (19), was not very successful for a time. It consisted of a series of retorts heated by fires underneath, and all the distillate was run together. This product was used mainly for the preservation of wood. Many people were experimenting with the idea, however, so that by 1888, according to Clark (5), the process of destructive distillation as applied to wood (longleaf pine) was an established and growing industry in the Southern States.

During this period many patents were taken out and many variations in the method of distillation were tried. The products marketed were charcoal, tar, pitch, creosote oils, and light oils. The light oils, which contained turpentine and pine oil, were not refined. In fact, it was not until about 1900 (7) that wood turpentine came into commercial notice, and even then the samples contained so many impurities that they could be used only for certain purposes and could be sold only at a decided reduction on the price for gum turpentine. Since the market for these oils was limited, the makers turned to the specialty market and sold them for secret uses, so that very often it was the maker who had the best selling agency, not the best plant, that succeeded. French and Withrow (7) cite the case of one firm having 100,000 gallons of a certain oil which they could not sell at 5 cents a gallon, while another firm could not supply its customers at 18 cents a gallon with the same product, but would not buy from the first for fear of future competition. The oils were used for paints, varnishes, stains, insecticides, disinfectants, medicinal products, and numerous other things.

After 1902 (23), when the steam-distilled wood turpentine came on the market, the destructive wood turpentine was also more thoroughly refined. Indeed, so much of the unpleasant odor was eliminated that in many instances it could be made to take the place of gum turpentine. About the same time the pine oil was also refined to a higher degree, and came to be commercially known as such, although it is still marketed also under various trade names.

At present the products of distillation are more or less standardized, but the plants vary in minor details. The capacity of the retorts varies from 1 to 11 cords. The retorts may be cylindrical or rectangular in cross-section and either horizontal or vertical. Some of them are heated by fires directly underneath and some by hot gases drawn through flues in the retort. The retorts also are heated to a higher temperature during the distillation in some plants than in others. The details depend to a large extent on the ideas of the man in charge and on what he considers to be his most important product. If it is charcoal, he will heat longer and at a higher temperature; if it is wood turpentine, he will heat carefully at first; and so on.

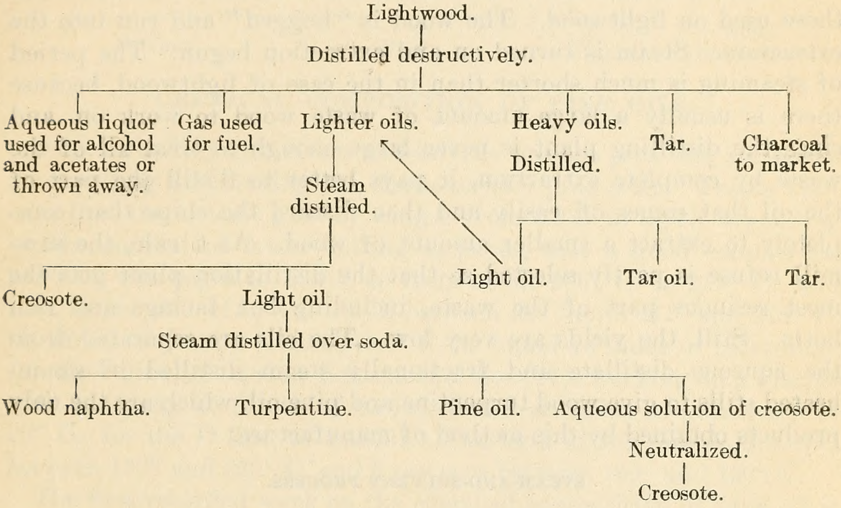
The methods in use in the spring of 1919 were in general as follows: The lightwood is brought to the plant in cordwood lengths, or shorter, and split to about the size of ordinary cordwood. The stumpwood, of course, is very crooked and does not pack well, but is usually richer in resinous material than the upper timber. This wood may be packed in the retort by hand as received, or after being cut into two-foot lengths, or it may be loaded on cars which are run into the retorts. The retorts are then sealed, steam run in to displace the air, and firing is begun. At first a mixture of aqueous distillate and light oils comes over. This contains most of the turpentine and pine oil, and the heat is kept low until it is over. Then the receiver is changed and the heat increased for the destructive part of the distillation. A fairly large amount of gas is formed in this part of the operation and is usually employed for firing. The heavy oils, which contain most of the tar oils, are gathered in a tank. This material may or may not be separated into two fractions. The tar may be distilled from the retort or it may be allowed to run out from the bottom of the retort through a water seal. When the distillation, which takes from 18 to 48 hours, is finished, the retort is permitted to cool down so that the charcoal will not take fire when it comes into contact with the air. Some plants provide covers about the size of the retort into which the cars containing the charcoal are drawn, and any fire is smothered by shutting off the air. Thus, less time between charges is lost, and the retorts are heated to some extent for the next distillation. In other plants it is necessary to cool the retorts sufficiently to permit the removal of the charcoal by hand, or even to allow a man to go inside to repack the retort. This usually causes a decided loss of time in the distillation cycle.

The oils are refined by fractional distillation in steam stills, repeated once or more according to the purity of the product desired, and distilled once, at least, from over soda solution to remove the creosote oils. The products are a wood naphtha, apparently quite similar to rosin spirits, wood turpentine, pine oil, tar oil, tar, pitch, and creosote. Owing to the heavy demand for flotation oils in the past few years, however, a very large amount of the oil has been sold unrefined for this purpose.

The yields of oils vary with the amount of resinous material in the wood and the methods employed in the distillation. Another factor in the reported variations of yields is that results are usually reported as so much a cord, while the wood charge is weighed rather than measured, the weight called a cord varying from about 3,500 to 5,000 pounds. Yields of total crude oils claimed vary from 40 to

120 gallons a cord, and the yield of pure pine oil was given as 1 to 4 gallons a cord. Veitch and Donk (28) give, for a cord of 4,000 pounds, a yield of from 36 to 120 gallons of crude oils and from 2 to 5 gallons of pine oil. At present the lower value may be partially due to the fact that the distillers must use poorer grades of lightwood than was necessary heretofore.

The following diagrammatic scheme shows the products of the destructive distillation of lightwood.



STEAM DISTILLATION.

Steam distillation of wood for turpentine and pine oil came later than the destructive distillation. Patents for this process were issued as early as 1864, but, according to Teeple (23), its rapid development did not begin until 1902, probably because of the fact that the price of turpentine, the main product, did not reach paying levels until that time.

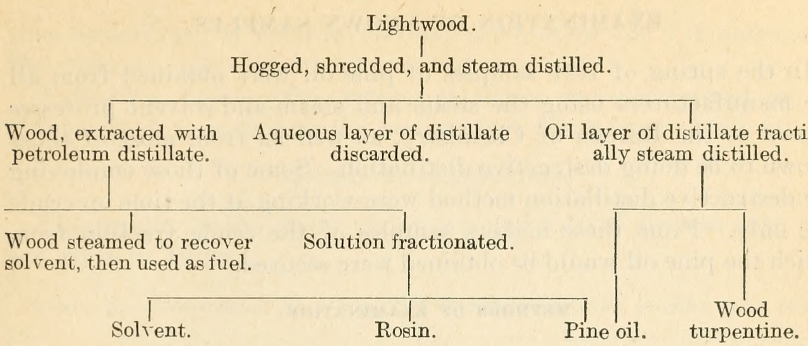
Many variations of the steam process, including the use of superheated and saturated steam, various pressures, various shapes of retorts, and different methods of steam circulation were patented; but finally practically the same method of operation was universally adopted (23). The wood is "hogged" (chipped), sometimes shredded, put in retorts, and subjected to a saturated-steam extraction. At the outset little or no pressure is used on the steam. When the volatile products decrease, the pressure is raised 10 or 15 pounds and subsequently reduced, when the steam and volatile products come out from the chips together. This increase and reduction of pressure is repeated several times. A yield of from 10 to 17 gallons a cord is obtained.

Probably the first wood turpentine sufficiently refined to compete with gum turpentine was put out by these plants. They are also to be credited with early refining of pine oil. By 1908 they were selling it in tank-car lots (20). Most of the steam distillation plants, however, were unable to exist on the low turpentine market of 1911–12, so that by 1913 no plants were operating, except one or two on sawmill waste. In the spring of 1919 none were known to be in operation other than in connection with sawmills.

The methods used on sawmill waste are somewhat different from those used on lightwood. The wood is "hogged" and run into the extractors. Steam is turned on and extraction begun. The period of steaming is much shorter than in the case of lightwood, because there is usually a large amount of waste wood to work on, and since the distilling plant is never large enough to treat all of the waste by complete extraction, it pays better to distill the part of the oil that comes off easily and then discard the chips than completely to extract a smaller amount of wood. As a rule, the sawmill refuse is partly selected so that the distillation plant gets the most resinous part of the waste, including box facings and rich butts. Still, the yields are very low. The oils are separated from the aqueous distillate and fractionally steam distilled in steam-heated stills to give wood turpentine and pine oil, which are the only products obtained by this method of manufacture.

STEAM-AND-SOLVENT PROCESS.

As the rosin is left in the waste wood by the steam process the problem of recovering it arises. The wood, being already shredded, is in good condition for extraction, and so the process of extraction with a volatile solvent is added to the steam process. As carried out at present, the lightwood is first "hogged," then shredded, and put into large extractors. Steam is turned on and steam distillation continued as long as an appreciable amount of oils comes over. Then the steam is turned off, a light petroleum distillate turned in, and extraction with this continued for some time with heating. When practically all the rosin is extracted the solvent-rosin mixture is run into a storage tank, and the chips are steamed to recover residual solvent, after which they are used for fuel. The oil from the aqueous layer is fractionally distilled into wood turpentine and pine oil. The solvent-rosin mixture is fractionated into solvent, pine oil, and rosin. The pine oil obtained from this process contains small amounts of the solvent, and for most commercial purposes it is sold mixed with the pine oil obtained from the steam part of the process. A diagrammatic scheme of the process is as follows:


Lightwood.
Hogged, shredded, and steam distilled.
Wood, extracted with petroleum distillate.
Aqueous layer of distillate discarded.
Oil layer of distillate fractionally steam distilled.
Wood steamed to recover solvent, then used as fuel.
Solution fractionated.
Solvent.
Rosin.
Pine oil.
Wood turpentine.

CHEMICAL COMPOSITION OF PINE OILS.

Steam-distilled and steam-and-solvent process pine oils are pleasant-smelling oils, having the characteristic pine odor. Teeple (20) says that the specific gravity ranges from 0.935 to 0.947, depending on the content of lower boiling terpenes, that a good commercial product will begin to distill at from about 206° C. to 210° C., and that 75 per cent of it will distill between the limits 211° and 218° C. On an oil of density 0.945 at 15.5° C. he found an index of refraction, for the D line, of 1.4830. Schimmel & Co. (17), working on a sample with a density at 15° C. of 0.9536, found an index of refraction at 20° C., for the D line, of 1.48537. The bulk of the sample distilled between 190° and 220° C. and 5 per cent between 160° and 190° C.

The first recorded work on the chemical composition of pine oil is that of Walker, Wiggins, and Smith (30), who concluded that this oil is a terpineol. Teeple (20) showed that the essential constituent of pine oil is a terpineol, probably alpha terpineol. Schimmel & Co. (17) report the following constituents: Camphene, alpha pinene, beta pinene, l-limonene, dipentene, cineol, gamma terpinene, alpha terpineol in large proportions, borneol, methyl chavicol, fenchyl alcohol, and camphor. The first seven of these, all of which boil at 180° C. or lower, doubtless are normal constituents of wood turpentine, and are found in pine oil because of incomplete separation. The last five have boiling points higher than 200° C. and are, therefore, probably normal constituents of pine oil.

The pine oil from destructive distillation varies much more than that from these processes. Chemically it is presumably a mixture of the oil described in the preceding paragraphs with oils formed by the destructive distillation of rosin and wood. The relative proportions of the two parts will vary with the process of distillation. In general, the distillation takes place over a somewhat wider range than in the case of the steam-distilled product.

EXAMINATION OF KNOWN SAMPLES.

In the spring of 1919 samples of pine oil were obtained from all the manufacturers using the steam and steam-and-solvent processes known to the Bureau of Chemistry, as well as from most of those known to be doing destructive distillation. Some of those employing the destructive distillation method were working at the time on crude oils only. From these makers samples of the crude fraction from which the pine oil would be obtained were secured.

METHODS OF EXAMINATION.

Specific gravity.—The specific gravity was taken with a Westphal balance at 20° C.

Distillation.—Distillation was conducted in an ordinary 250 cc. distilling flask, using 50 cc. samples, and was at the rate of about one drop a second. The temperatures are uncorrected.

Resinous material.—In the case of the pine oil the content of resinous material was very small, and it was determined by shaking out the sample with two portions of 25 per cent sodium hydroxid solution, washing the solution twice with ether, acidifying the alkaline extract with dilute sulphuric acid, extracting the resinous material with ether, taking down on the water bath, and weighing. In the case of the wood naphtha and the crude distillates it was determined as directed under " Phenols."

Phenols.—The total alkali-soluble material, extracted as described under " Resinous material," was steam distilled after acidifying with dilute sulphuric acid, and the distillate saturated with salt and extracted with ether. The ether solution was taken down on the water bath and weighed as phenols. The residue in the distillation flask was shaken out with ether, the ether evaporated off, and the residue weighed as resinous material. With ferric-chlorid solution, all the phenols gave a blue coloration, changing to brown on standing. With solid potassium hydroxid and chloroform, they gave a reddish-purple color, indicating the presence of guaiacol.

Unpolymerized residue.—Twenty cubic centimeters of exactly 38 N sulphuric acid, in a graduated narrow-neck Babcock bottle, were cooled in ice water; 5 cc. of the oil to be examined were slowly added and mixed, care being taken that the temperature was kept below 60° C. When the mixture no longer warmed on shaking, it was thoroughly agitated, the bottle was placed in a water bath heated to from 60° to 65° C. for 10 minutes, and the contents were thoroughly mixed by vigorous shaking six times, for one-half minute each time, during the period. The bottles were then cooled to room temperature and filled with concentrated sulphuric acid until the contents came up to the graduations in the necks of the bottles. They were then centrifuged at 1,200 revolutions per minute, for 5 minutes, and the volume of unpolymerized material read off.

Refractive index.—The refractive index was determined on an Abbé type refractometer, the temperature being kept at 20° C.

RESULTS OF EXAMINATION.

The results on the steam and steam-and-solvent process pine oils are given in Table 1.

TABLE 1.—*Composition of steam and steam-and-solvent process pine oils.*

Laboratory No.	Method of manufacture.	Density at 20° C.	Refractive index at 20° C.	Water.	Oil distilled below 190° C.	Distillate, 190°-220° C.	Unpolymerized.	Resinous material.
				Per cent.	*Per cent.*	*Per cent.*	*Per cent.*	*Per cent.*
31704	Steam	0.932	1.4870	Trace.	3.0	87.6	2.0	0.09
31859	do	.942	1.4848	0.6	1.8	94.6	1.2	.37
31860	do	.945	1.4833	.8	1.2	92.0	1.2	.05
31705	Steam-and-solvent	.930	1.4820	1.0	3.6	92.0	1.2	.17
31706	do	.927	1.4820	.6	7.0	89.0	2.0	.27
31707	do	.932	1.4835	.4	4.0	92.4	3.2	.20

These oils, although obtained from widely separated points, are quite similar according to these analyses. The densities vary from 0.927 to 0.945, and the refractive indices from 1.4820 to 1.4870. In no case did the water exceed 1 per cent or the resinous material 0.5 per cent. The distillate between 190° and 220° C. was from 87.6 to 94.6 per cent.

The destructive-distillation pine oils vary much more in their properties, as will be seen from the results in Table 2.

TABLE 2.—*Composition of destructive-distillation pine oils.*

Laboratory No.	Density at 20° C.	Refractive index at 20° C.	Water.	Oil distilled below 190° C.	Distillate 190°-220° C.	Unpolymerized.	Resinous material.
			Per cent.	*Per cent.*	*Per cent.*	*Per cent.*	*Per cent.*
31708	0.893	1.4868	Trace.	47.0	50.2	0.8	0.02
31709	.903	1.4920	Trace.	14.8	82.0	.8	.38
31711	.929	1.5000	None.	None.	98.0	.8	.10
31712	.949	1.4995	0.4	Trace.	91.4	.8	.10
31713	.944	1.5035	Trace.	11.0	65.4	.8	[1] 12.50
31714	.886	1.4945	Trace.	65.2	32.4	.8	[1] 1.70

[1] Contained appreciable amounts of wood-tar phenols or creosote.

The densities vary from 0.886 to 0.949; the refractive indices from 1.4868 to 1.5035; the distillate below 190° C., from none to 65.2 per cent; and the alkali-soluble material from 0.02 to 12.5 per cent. In color the oils vary from a light yellow, similar to that of steam-process pine oil, to a dark reddish brown. Certain of them are almost free from the smoky odor characteristic of destructive wood distillates, while others are heavy with it.

Since it was desired to determine whether other oils from the destructive distillation of wood are active germicides, commercial samples of these also were collected and the samples analyzed (Table 3).

TABLE 3.—*Composition of commercial samples of destructive-distillation pine oils.*

Laboratory No.	Description of sample.	Density at 20°C.	Refractive index at 20°C.	Resinous material.	Phenols.	Water.	Oil distilled.			Unpolymerized.
							Below 160° C.	160° C. to 180° C.	180° C. to 220° C.	
				Per ct.	*Per ct.*	*Per ct.*	*Per ct.*	*Per ct.*	*Per ct.*	*Per ct.*
31710....	Wood naphtha....	0.869	1.4780	[1] 2.59	....	None.	34.0	56.0	6.0	0.8
31715....	.....do..............	.869	1.4625	2.07	0.54	None.	83.0	12.6	....	.8
31716....	.....do..............	.868	1.4700	.74	2.43	None.	79.0	16.0	....	.4
31717....	Crude light oil.....	.919	1.4965	11.90	3.85	None.	12.0	39.6	21.0	Trace.
31720....	.....do..............	.909	1.4840	2.96	5.30	None.	25.0	39.6	28.4	.8
31723....	.....do..............	.946	1.5105	19.70	4.40	None.	4.0	31.0	27.0	.4
31719....	Heavy crude oil...	1.028	1.5450	37.40	6.20	0.6	4.0	8.0	16.0	None.
31718....	Entire crude oil...	.975	1.5255	20.10	5.80	None.	19.4	14.6	12.8	0.8
31721....	.....do..............	1.020	1.5460	37.90	4.80	1.0	6.0	8.0	15.0	Trace.
31722....	.....do..............	1.003	1.5355	26.80	6.40	.8	6.4	10.0	20.0	Trace.
31725....	Tar oil..............	1.049	1.5450	31.30	9.70	1.0	1.6	1.0	8.0	None.
31726....	.....do..............	1.015	1.5500	22.60	6.00	.4	2.0	6.0	18.0	Trace.

[1] Contained wood-tar phenols which were not separated.

It will be noted that there is a rough relation between the boiling points, densities, and indices of refraction of the crude oils, those with the lower boiling points having the lower densities and lower refractive indices. The unpolymerized residue is in all cases less than 1 per cent. The resinous material and phenols are high in the crude distillate, in some cases as much as 40 per cent.

Pine oil obtained by the steam or steam-and-solvent process is a very uniform product. That obtained from destructive distillation is much less uniform. It consists of the same compounds as the steam-distilled product, together with the distillation products of rosin and wood in various proportions, depending upon the particular manner in which the distillation was conducted.

The crude distillation products also vary greatly in their composition, depending on the practice at the plant where they were made.

The unpolymerized residue in the case of all the products of pine distillation is low, 2 per cent or less, except in one case in which the oil had been in contact with petroleum distillate in the course of manufacture and probably still contained a small amount of it. Since petroleum oils, of course, give high polymerization residues, this provides a method for detecting the adulteration of pine oil with kerosene or other mineral oil fractions.

PREPARATION OF PINE-OIL EMULSIONS.

All of the samples collected, which were fairly representative of the products of destructive distillation of the longleaf pine as carried out commercially, were made into emulsifiable liquids by the method given for the Hygienic Laboratory pine-oil disinfectant (18), with certain modifications in some cases.

The oil, rosin, and alkali solutions were mixed in the following proportions and according to the following directions:

	Grams.
Oil	1,000
Rosin	400
25 per cent sodium hydroxid solution	200

The oil and rosin were heated together in a covered porcelain vessel until all the rosin dissolved. The mixture was cooled to 80° C., the sodium hydroxid solution added, and the liquid violently stirred for at least 10 minutes. Sufficient water was added to make the mixture to the original weight, and the preparation cooled. It will be noted, of course, that only the steam or steam-solvent oils are used in the Hygienic Laboratory pine-oil disinfectant. Oils Nos. 31717, 31718, 31719, 31725, and 31726 already contained so much resinous material that they did not emulsify well when the full amount of rosin called for by the Hygienic Laboratory formula had been added to them. Consequently, less rosin was used in these cases, one-half the specified amount in the case of Nos. 31717 and 31718, and no additional rosin in the others. The usual amount of sodium hydroxid solution was added in these mixtures. In mixing with water all gave fair emulsions.

These products were then used for determining the bactericidal efficiency, the results of which are given in the following pages.

DISINFECTANT ACTION OF EMULSIONS OF PINE-OIL AND OTHER PINE-DISTILLATION PRODUCTS.

STEAM-DISTILLED AND STEAM-AND-SOLVENT PROCESS PINE-OIL EMULSIONS.

Stevenson (18) claimed for a pine-oil emulsion made from steam or steam-and-solvent process pine oil, advocated by him in 1915 as a general disinfectant, a phenol coefficient of from 4 to 6, and stated that it could be used in any dilution up to $\frac{1}{500}$, according to the time it was allowed to act. The dilution recommended for practical use seems to have been based solely on the Hygienic Laboratory phenol coefficient. Stevenson also stated that the preparation deteriorates with age. Later McCoy, Stimson, and Hasseltine (13), and Walters (31) called attention to the fact that this pine-oil disinfectant, although showing a high disinfectant value against organisms of the typhoid group and certain others, is much less efficient than phenol or cresol against other common pathogenic organisms, notably *Micrococcus aureus.*

In order to check these various statements, six samples of pine oil were prepared according to the directions given on page 11 of this bulletin, and their Hygienic Laboratory phenol coefficients determined immediately after making, at the end of a month, and in from 12 to 13 months. In these tests Armour's peptone was substituted for Witte peptone, which was difficult to obtain and not of uniform quality. The same lot of peptone was used throughout, but the meat extract used in the broth was from two different batches, the one employed at the end of the experiments being much darker than the other. The tests were made by two operators using the same materials.

When freshly made, these six samples gave coefficients varying from 3.42 to 4.34. At the end of a month the same samples gave coefficients ranging from 3.31 to 4.19, and at the end of 12 months they gave coefficients ranging from 3.14 to 4.23. The greatest single variation obtained was a drop from 3.85 when freshly made to 3.14 at the end of a year. The average of the coefficients obtained on these six freshly made preparations was 3.88. At the end of the month the average was 3.74, and at the end of 12 months the average was 3.66. This would seem to indicate a deterioration of less than 10 per cent in one year. Taking into consideration the limitations of the test, however, it is evident that this apparent deterioration is practically negligible.

With respect to dilutions, none of the samples examined were uniform in killing *B. typhosus* in $\frac{1}{500}$ dilution in 15 minutes, and two failed to kill in $\frac{1}{400}$ dilution in this length of time. While it is probably true that a $\frac{1}{500}$ dilution would have killed if allowed to remain in contact with the organism for an indefinite time, yet, considering the difficulties of practical disinfection as compared to the artificial test, it is evident that a $\frac{1}{500}$ dilution is too high to recommend even for *B. typhosus*. Under the circumstances it would be much safer to recommend a dilution of not over 1 in 100 when used in cases of typhoid fever.

In testing the efficiency of the product against *Micrococcus aureus*, several strains of this organism were employed, as it varies markedly in its resistance to pine-oil preparations. In all, four strains were tested. These were given laboratory numbers 200, 202, 203, and 204. No. 200, a stock strain over five years old, formerly used for class demonstrations, at that time gave typical lesions in the rabbit. At present it is practically nonpathogenic. Nos. 202 and 203 were freshly isolated by the writer from infections in the finger and neck. No. 202 caused abscess formation in the rabbit. No. 203 was nonpathogenic for this animal. No. 204 was a freshly isolated strain obtained from the United States Navy medical school through the courtesy of Dr. Pryor. It killed the rabbit, when injected intravenously in dosage of 2 cc. broth culture, in from one to two days. The lesions produced were those of septicæmia. All the strains produced pigment on potato, and microscopically and culturally seemed typical. They could be differentiated one from the other only by animal inoculation and by the difference in resistance shown by one of them to pine oil.

In testing the pine-oil emulsions against these strains the Rideal-Walker technique was used, and, in order to guard against possible error through contamination, the pathogenic strains were, in at least one test, passed through a rabbit after recovery from the pine oil.

No. 200, the old stock strain, was found to be the most variable in its power of resistance. It was tested against all six samples of the Hygienic Laboratory pine-oil disinfectant, and gave R. W. coefficients ranging from none to 1. In no case, however, was it killed in 15 minutes by a dilution of $\frac{1}{100}$. The variability of this strain made it obviously unsuited for experimentation, for which reason the other three strains were obtained.

Nos. 202, 203, and 204 acted alike, and were found to resist 5 and 10 per cent emulsions for 15 minutes. Since 10 per cent is too strong to make a satisfactory emulsion, it was evident that the product is, for all practical purposes, useless against these strains in any strength.

In order to test the efficiency of pine oil against the spore-bearing group of bacteria, a number of experiments were made in which *B. anthracis* was employed as the test organism. Old neutral-agar cultures were used. To these sterile distilled water was added, after which small pieces of sterile filter paper were saturated in the resulting mixture. These pieces of paper were transferred to test tubes containing 5 and 10 per cent dilutions of the pine-oil emulsion. After various intervals of time they were again transferred to tubes of broth, in order to free them from the pine oil, and finally transferred to neutral agar. The tubes were kept at a room temperature of from 20° to 25° C. Two samples were tested in 10 per cent emulsion and one in 5 per cent emulsion. Under these conditions pine oil failed to kill the spores of *B. anthracis* in 3 days.

EMULSIONS OF DESTRUCTIVE-DISTILLED PINE OIL AND OTHER PRODUCTS OF DISTILLATION.

In addition to the sample of Hygienic Laboratory pine-oil disinfectant prepared by the method described by Stevenson, there are on the market a number of preparations made from inferior oils. These (page 9) are known as destructive-distilled pine oil, wood naphtha, crude light oil, entire crude oil, and tar oil. Experiments on these preparations were carried out in the same way as those on the Hygienic Laboratory pine-oil disinfectant.

Five samples of the destructive-distilled pine-oil emulsions gave coefficients ranging from 1.71 to 3.42 when freshly prepared. At the end of a month the range was from 1.68 to 3.40, and at the end of the year from 1.45 to 3.23. The greatest individual drop in one year was from 1.71 when freshly made to 1.45 at the end of the year. The average was 2.47 when freshly prepared, 2.52 at the end of the month, and 2.46 at the end of the year. No deterioration could be demonstrated. It should be noted that these samples varied one from the other more markedly than did the Hygienic Laboratory pine-oil disinfectants, one showing a coefficient of 1.71, another a coefficient of 3.42.

Two samples of the wood naphtha oil emulsion were examined. One gave a coefficient of 0.70 when freshly prepared, 0.60 at the end of a month, and 0.60 at the end of a year. The other gave a coefficient of between 0.54 and 0.60 when fresh, 0.44 at the end of a month, and 0.45 at the end of a year. The slight decrease in the coefficient noted at the end of the first month is too small to indicate any definite deterioration.

A sample of "light" crude-oil emulsion gave a coefficient of 0.85 when fresh, 0.75 at the end of a month, and 0.90 at the end of the year. A second sample, examined within a month of its manufacture, showed a coefficient of 0.84. At the end of the year the coefficient was 1.03.

A sample of the "heavy" crude-oil emulsion had a coefficient of 0.74 when examined in the first month. At the end of a year it was 0.78.

A sample of emulsion made from the entire crude oil gave a coefficient of 0.60 when fresh, 0.64 at the end of the month, and 0.64 at the end of the year. A second sample gave a coefficient of 0.74 at the end of a month, and 0.85 at the end of the year.

Two samples of tar-oil emulsions showed coefficients of 0.82 and 0.70 in the first month, and 0.84 and 0.83, respectively, at the end of a year.

The action of these samples on *M. aureus* and *B. anthracis* was similar to that of the Hygienic Laboratory pine-oil disinfectant. That is to say, when tested against *M. aureus* strain 200, they gave Rideal-Walker coefficients ranging from none to 0.7, but when tested against strains 202 and 203, they failed to kill in any dilution. Likewise they failed to kill *B. anthracis* in 5 and 10 per cent emulsions.

CONCLUSIONS.

Pine-oil emulsions made from steam-distilled pine oils, when freshly prepared, gave Hygienic Laboratory coefficients varying from 3.42 to 4.34, the average being 3.88. At the end of 12 months the average was 3.66.

A disinfectant prepared from destructive-distilled pine oil is weaker as well as more variable in its germicidal power against *B. typhosus* than is the Hygienic Laboratory pine-oil disinfectant. The samples examined gave Hygienic Laboratory coefficients of from 1.71 to 3.42.

Emulsions made from the other oils tested gave coefficients under 1. These preparations failed to emulsify completely in 10 per cent concentration.

Pine-oil emulsions made from various grades of pine oils failed to kill *M. aureus* and *B. anthracis* in any dilution capable of emulsification.

In view of the results obtained these products should not be used for general disinfecting purposes.

When using pine-oil emulsions against *B. typhosus*, it is safer, for practical purposes, to employ a solution of five times the strength capable of killing the organism in five minutes. Thus a product showing by the Hygienic Laboratory method a killing power of $\frac{1}{500}$ should be used in a $\frac{1}{100}$, or 1 per cent, dilution. If a product will not give a dilution having a concentration five times that of the weakest concentration capable of killing *B. typhosus* in 15 minutes, and remain completely emulsified, it should not be used as a disinfectant.

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